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A NEW SUPERCONDUCTING MATERIAL IN A Y-Ba-Cu-O AND Bi-Ca-Sr-Cu-O MIXED SYSTEM

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A new superconducting phase has been found in a mixed system prepared from the $YBa_2Cu_3O_{7-\delta}$ and $Bi_2CaSr_2Cu_2O_{9-\delta}$ high-T_c materials. The new compound shows an onset of resistivity transition at ~100 K and zero resistivity below ~50 K. Powder X-ray diffraction pattern of the new compound differs completely from those of $YBa_2Cu_3O_{7-\delta}$ and $Bi_2CaSr_2Cu_2O_{9-\delta}$. The materials prepared using a mixture of the oxides and carbonates with identical cation stoichiometric ratios, on the other hand, does not exhibit superconductivity.

1. Introduction

The discovery of high temperature superconductivity by Bednorz and Müller[1] has resulted in finding a number of new oxide systems with T_c above 77 K.[2-6] It is also interesting to search for superconductivity in materials consisting of mixture of these high-T_c oxides. To our knowledge, no such work has been reported so far. There is also the possibility that a higher T_c can be achieved with such multi-cation mixture. In this study, we have observed a new superconducting phase in a mixture prepared from $YBa_2Cu_3O_{7-\delta}$ and $Bi_2CaSr_2Cu_2O_{9-\delta}$ high-T_c materials.

2. Experimental

Y_2O_3, Bi_2O_3, $BaCO_3$, $CaCO_3$, $SrCO_3$, and CuO, all of 99.9 % purity were used as starting materials. $YBa_2Cu_3O_{7-\delta}$ samples were prepared by mixing, dry grinding and firing appropriate quantities at 950°C for 12 hours in flowing oxygen, followed by sintering the pellets under identical conditions. $Bi_2CaSr_2Cu_2O_{9-\delta}$ samples were prepared by mixing the starting materials in the cation ratio Bi:Ca:Sr:Cu = 1:1:1:2, grinding and firing the powder at 850°C in air for 12 hours followed by sintering the pellets at 860°C in air for 4 hours. The two superconducting materials were characterized by resistivity, Meissner effect and X-ray powder diffraction measurements. The mixed system A was then prepared by mixing the two superconductors in the 1:1 cation molar ratio, and sintering the pellets at 950°C for 12 hours in flowing oxygen. Resistivity of the pellet samples was measured in a closed cycle refrigerator, model Daikin V202A5L using the four-probe technique with pressed indium contacts. The sample temperature was measured by a gold-chromel thermocouple. Meissner effect was demonstrated by observing the deflection of a vertically suspended small samarium-cobalt magnet. X-ray powder diffraction measurments were performed using a Shimadzu model XD-7A X-ray diffractometer using Cu-Kα radiation. Compound *B* was prepared by using the individual oxides or carbonates as the starting materials instead of the Y and Bi superconductors. Oxides were mixed in the same cation ratio as for the mixed system *A* and sintered at 950°C, pelletized and sintered again at 950°C for 12 hours. Sintering of the pellets was repeated four times, in order to have a similar annealing conditions for both systems *A* and *B*.

3. Results and Discussion

X-ray powder diffraction patterns of $YBa_2Cu_3O_{7-\delta}$ and $Bi_2CaSr_2Cu_2O_{9-\delta}$ superconductors are shown in Fig. 1. They are in agreement with the data reported in literature.[4,5,7,8] Resistance versus temperature curves for the Y and Bi superconductors are shown in Figs. 2 and 3. Y–compound has an onset of resistivity transition at 95 K and zero resistivity at 85 K. The Bi–compound exhibits multiphase behavior having the onsets of resistivity transition at 115 K

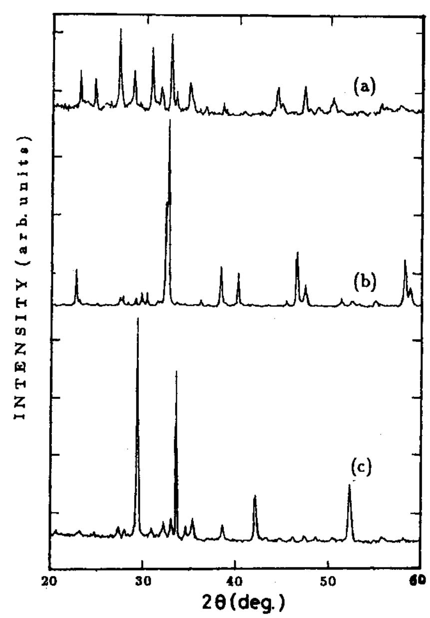

Fig. 1. The XRD patterns of (a) $Bi_2CaSr_2Cu_2O_{9-\delta}$ (b) $YBa_2Cu_3O_{7-\delta}$ and (c) mixed system *A* prepared from Y and Bi superconductors.

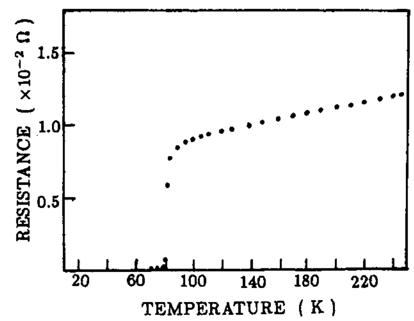

Fig. 2. Temperature dependance of the resistance for $YBa_2Cu_3O_{7-\delta}$.

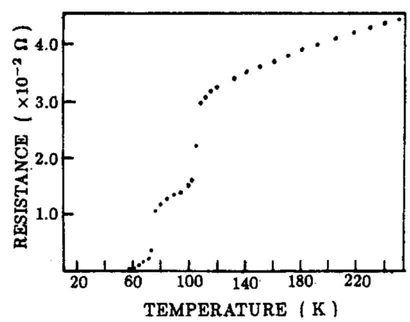

Fig. 3. Temperature dependence of the resistance for $Bi_2CaSr_2Cu_2O_{9-\delta}$.

and 80 K, respectively, and zero resistivity at 60 K. The onset of the Meissner signal was observed in the Y sample at 95 K and in the Bi sample at ~115 K.

Figure 4 shows (a) the XRD pattern of the mixed system *A* prepared from the Y and Bi superconductors and (b) the XRD pattern of the mixed system *B* prepared from the oxides/carbonates with identical cation ratios and after four heat treatment cycles. XRD pattern of *A* differs completely from the XRD patterns of individual Y and Bi superconductors shown in Fig. 1, suggesting the existence of a new phase or phases in system *A*.

The resistance versus temperature curves for systems *A* and *B* are shown in Figs. 5 and 6, respectively. System *A* exhibits superconducting behavior with an onset of resistivity transition at ~100 K and zero resistivity at ~55 K. System *B* has a rather high room temperature resistance and exhibits semiconducting behavior with the decrease in temperature. Meissner effect was observed with system *A* starting around 100 K whereas no such effect was observed with system *B* down to ~15 K. Although the XRD patterns of these two systems appear to be similar in their broad features and principal peaks, noticeable differences can be seen in the finer details of the few main diffraction peaks. Comparison of the

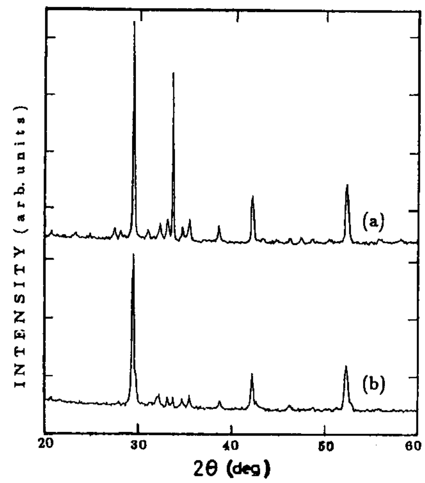

Fig. 4. The XRD patterns of (a) mixed system *A* prepared from the Y and Bi superconductors and (b) mixed system *B* prepared from the oxides/corbonates with identical cation ratios.

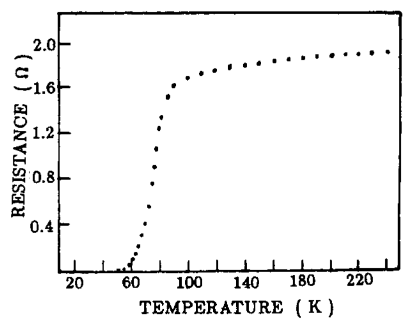

Fig. 5. Temperature dependence of the resistance for the mixed system *A* prepared from the Y and Bi superconductors.

XRD peaks of *A* and *B* at $2\theta = 29.5°$, $42°$, and $52.5°$ clearly shows the existence of an additional shoulder at each of these peaks in the XRD pattern of *B*. Also the peak at $2\theta = 34°$ is much more intense for *A*. These finer differences in the XRD patterns of systems *A* and *B* suggest that the structures and compositions of different phases in *A* and *B* are different. In the present case, it is also possible that *A* and *B* are two different phases of the six component mixture where only *A* is superconducting and this phase, perhaps unstable over long periods, is not achievable by the procedure adopted to prepare *B*. The finer differences in the XRD patterns of *A* and *B* could also be due to the presence of impurity phases in *B*. Such impurity phases at grain boundaries could mask the transport properties of superconducting grains and could give rise to semiconducting behavior in the

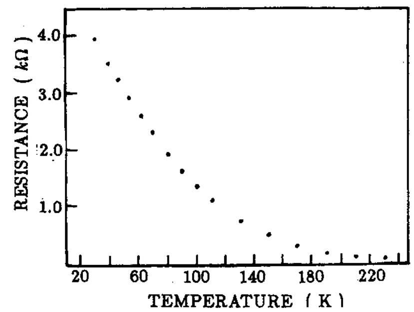

Fig. 6. Temperature dependence of the resistance for the mixed system *B* prepared from the oxides/carbonates with identical cation ratios.

bulk even if both *A* and *B* consist of isostructural bulk phases. However, the absence of Meissner effect in *B* rules out this possibility. It can, therefore, be concluded that either *A* and *B* are two different products, probably with multiphase structure or two different phases of the same compound.

Our observations reveal an important experimental finding which may be useful in the preparation of high temperature superconductors. High-T_c materials are generally prepared by the solid state reaction of appropriate metal oxides/carbonates. When this technique is applied to systems with several oxides, the final product may depend on the route followed, as evidence by the present investigation.

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